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(54) Actinide dissolution

(57) An actinide such as plutonium is dissolved at a pH of 5.5 or greater by treating the actinide with a conditioning agent and a complexing agent in an aqueous medium. For example the conditioning agent may comprise hydrogen peroxide and the complexing agent may comprise citrate ions. The pH of the aqueous medium is controlled using carbon dioxide or a carbonate such as sodium carbonate to above 5.5. The method is particularly useful in the dissolution of Magnox-sheathed fuel elements.

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The claims were filed later than the filing date within the period prescribed by Rule 25(1) of the Patents Rules 1982.

Actinide Dissolution

This invention relates to the dissolution of actinides and is particularly, but not necessarily exclusively concerned with the dissolution of nuclear fuel.

According to the present invention there is provided a method of dissolving an actinide or fission product, said method comprising contacting the actinide or fission product with a conditioning agent, a complexing agent and an aqueous medium, and controlling the pH of the aqueous medium such that the pH is maintained above 5.5 and so that the actinide or fission product dissolves in the aqueous medium.

The term fission product as used herein refers to those products formed in the fission of nuclear fuel.

Preferably, the actinide comprises plutonium or a plutonium-containing mixture of actinides. The actinide may also comprise uranium. The actinide may be derived from nuclear fuel and in particular irradiated nuclear fuel, and may comprise a compound of the actinide such as an oxide, hydroxide or carbonate.

The pH of the aqueous medium may be controlled using carbon dioxide and/or a carbonate such as sodium carbonate. For example, carbon dioxide may be passed into the aqueous medium to give a pH of about 5.5 to 10,

or sodium carbonate may be added to give a pH of about 10 or above.

The actinide or fission product may dissolve in the aqueous medium as a carbonate compound or complex.

The pH of the aqueous medium may be neutral or near neutral.

The complexing agent may comprise the anion of a carboxylic acid such as citric acid, acetic acid, formic acid, oxalic acid or ethylenediamine tetra-acetic acid.

The conditioning agent may comprise an oxidising agent such as hydrogen peroxide, ozone, oxygen-enriched air or potassium permanganate, or a reducing agent such a hydrazine or hydroxylamine.

The actinide or fission product may be present on a surface of a component, such as a concrete component, and the method used to decontaminate the surface.

The actinide or fission product may be present in a medium based on magnesium hydroxide and the method used to dissolve the actinide and said medium. For example, the actinide may comprise plutonium dioxide and/or uranium dioxide, the medium may comprise an alloy known as Magnox (a family of magnesium alloys containing traces of aluminium, manganese and zirconium) and its corrosion products (mainly magnesium hydroxide), and carbon dioxide used to control the pH so that magnesium is dissolved as its bicarbonate.

It is believed that the conditioning agent changes the oxidation state of the actinide (or fission product)to a state which renders the actinide (or

fission product) soluble by forming a complex with the complexing agent or by the natural solubility of the conditioned cation in the leaching solution. The complex produced by the complexing agent and the actinide (or fission product) may facilitate the oxidation or reduction carried out by the conditioning agent.

The invention will now be further described, by way of example only, with reference to the accompanying examples.

One type of fuel used in nuclear reactors comprises uranium metal which is housed within a metallic sheath typically made from Magnox (a magnesium alloy containing traces of aluminium, manganese and zirconium). When the irradiated fuel is removed from the nuclear reactor it is first allowed to cool and then separated from the Magnox sheath. The Magnox sheath and any fragments of irradiated nuclear fuel associated with the sheath are stored under water in concrete containers. Corrosion of the Magnox during prolonged storage gives a sludge containing Magnox metal, magnesium hydroxide, uranium dioxide and products generated in the irradiation of the fuel, eg plutonium dioxide, americium and fission products.

An aqueous suspension of the sludge was stirred, carbon dioxide gas passed into the sludge, and solutions of sodium citrate (0.01 to 0.05M) and of hydrogen peroxide (0.02M) added. A solution was obtained which was sent for further processing, such as ion exchange.

Passage of carbon dioxide into the sludge provides bicarbonate ions and magnesium is dissolved as magnesium bicarbonate. Following dissolution of the sludge the magnesium can be removed from solution by heating when the soluble bicarbonate is converted to insoluble carbonate. Other actinides, such as americium and neptunium, also dissolve in the carbon dioxide sparged aqueous solution containing the peroxide and citrate ions.

It is thought that the citrate and insoluble plutonium form a soluble plutonium complex, and that the presence of the citrate may facilitate the oxidation of the plutonium by the hydrogen peroxide to a more soluble oxidation state, such as plutonium (VI). A similar mechanism may operate with uranium and may assist the reduction of, for example, insoluble plutonium (IV) to soluble plutonium (III).

Since the dissolution is carried out at pH's above 5.5 and can be carried out at about neutral pH components such as concrete are not attacked. This contrasts with the usual methods of dissolving nuclear fuel which employ nitric acid - a reagent which attacks concrete.

The method of dissolution can be used to decontaminate components by immersing the components in an aqueous solution and passing carbon dioxide through the solution and adding citrate and hydrogen peroxide. To reduce loss of carbon dioxide from such a system it is

advantageous to carry out the dissolution in a closed vessel or to re-circulate the carbon dioxide. Alternatively the surface to be decontaminated can be contacted with a solution of hydrogen peroxide, sodium citrate and sodium carbonate by, for example, spraying the solution onto the surface.

When decontaminating concrete surfaces it is advantageous to employ sodium carbonate in place of carbon dioxide as a pH of about 10 can be maintained which reduces the amounts of salts present which can corrode the concrete.

Since magnesium provides anions to counter balance bicarbonate ions generated from the carbon dioxide, when the method is used in instances where magensium is not present an ion can be added to provide the balance.

Although the example described refers to the dissolution of Magnox sludge containing actinide derived from irradiated nuclear fuel the method may be used to dissolve actinides not so derived, for example in uranium ore processing for dissolving uranium and associated materials.

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Claims

1. A method of dissolving an actinide or fission product comprising contacting the actinide or fission product with a conditioning agent, a complexing agent and an aqueous medium, and controlling the pH of the aqueous medium such that the pH is maintained above 5.5 and so that the actinide or fission product dissolves in the aqueous medium.

2. A method as claimed in claim 1 which comprises controlling the pH of the aqueous medium using carbon dioxide.

3. A method as claimed in claim 1 or 2 which the conditioning agent comprises an oxidising agent or a reducing agent.

4. A method as claimed in claim 1, 2 or 3 in which the complexing agent is the anion of a carboxylic acid.

5. A method of dissolving an actinide or fission product as claimed in claim 1 substantially as herein described.

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